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13-12-2012	Final Report	19-Aug-2008 - 18-Aug-2012

4. TITLE AND SUBTITLE

Bright Coherent Optical Waveforms from the Infrared to the Vacuum Ultraviolet for Manipulation and Detection of Molecules

5a. CONTRACT NUMBER
W911NF-08-1-0391

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER
611102

6. AUTHORS

Margaret Murnane, David Jonas and Henry Kapteyn

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES

University of Colorado - Boulder
The Regents of the University of Colorado
Office of Contracts and Grants
Boulder, CO 80309 -0572

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

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11. SPONSOR/MONITOR'S REPORT NUMBER(S)
54364-PH.6

12. DISTRIBUTION AVAILIBILITY STATEMENT

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13. SUPPLEMENTARY NOTES

The views, opinions and/or findings contained in this report are those of the author(s) and should not contrued as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

In this grant we made four major advances in extreme nonlinear optics and strong field science. First we showed that it is possible to induce electromagnetic transparency in an atom (He) subject to intense vacuum VUV radiation. This allowed us to enhance or suppress multiphoton ionization of an atom by manipulating two interfering pathways for ionization. Second visualized in space and time the dynamic evolution of the entire ten-electron valence shell electron density as a molecular bond ruptures – a feat that was never before achieved. Third, we

15. SUBJECT TERMS

Bright Coherent Optical Waveforms from the Infrared to the Vacuum Ultraviolet for Manipulation and Detection of Molecules

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Margaret Murnane
UU	UU	UU	UU		19b. TELEPHONE NUMBER: 303-210-0396

Report Title

Bright Coherent Optical Waveforms from the Infrared to the Vacuum Ultraviolet for Manipulation and Detection of Molecules

ABSTRACT

In this grant we made four major advances in extreme nonlinear optics and strong field science. First we showed that it is possible to induce electromagnetic transparency in an atom (He) subject to intense vacuum VUV radiation. This allowed us to enhance or suppress multiphoton ionization of an atom by manipulating two interfering pathways for ionization. Second visualized in space and time the dynamic evolution of the entire ten-electron valence shell electron density as a molecular bond ruptures – a feat that was never before achieved. Third, we theoretically developed a new and generalized realization of quasi phase matching (QPM) - called spatiotemporal quasi-phase matching - that generalizes traditional spatial QPM using layered media to space-time QPM. Instead of simply being described by a momentum mismatch, in general a nonlinear optical process may possess an energy mismatch, a momentum mismatch, or both an energy and momentum mismatch. Finally, we developed a new understanding of high harmonic generation that allowed us substantially increased the conversion efficiency into the vacuum ultraviolet region of the spectrum by using UV and VUV driving lasers.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
10/05/2011	1.00	Alon Bahabad, Margaret Murnane, Henry Kapteyn. Quasi Phase Matching of Momentum and Energy in Nonlinear Optical Processes, Nature Photonics, (06 2010): 570. doi:
10/05/2011	2.00	Wen Li, Agnieszka Jaron-Becker, Craig Hogle, Vandana Sharma, Xibin Zhou, Agnieszka Becker, Henry Kapteyn Margaret Murnane. Visualizing electron rearrangement in space and timeduring the transition from a molecule to atoms, PNAS, (11 2010): 20219. doi:
10/05/2011	3.00	P. Ranitovic, X. M. Tong, C. W. Hogle, X. Zhou, Y. Liu, N. Toshima, M. M. Murnane, H. C. Kapteyn. Controlling the XUV Transparency of Helium using Two Pathway Quantum Interference, Physical Review Letters, (07 2011): 193008. doi:
TOTAL:	**3**	

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received		Paper
TOTAL:		

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Ahmed Zewail Prize talks, Annual Meeting of the American Chemical Society, Salt Lake City, UT March 2009. (Two talks presented by Margaret Murnane and Henry Kapteyn)
Invited talk, “Observing the Dance of Electrons in Atoms, Molecules and Materials using Coherent Electrons and x-rays,” Graduate Student Symposium, Division of Atomic, Molecular, and Optical Physics of the American Physical Society (DAMOP), Charlottesville, May 2009. Presented by Margaret Murnane.
Invited talk, “Harnessing Attosecond Science for Extreme Nonlinear Optics,” 18th International Laser Physics Workshop (LPHYS'09), Barcelona, Spain, July 2009. Presented by Margaret Murnane.
Invited talks, Femtochemistry, Femtobiology, and Femtophysics (Femtochemistry IX), Beijing, China, August, 2009. (Invited talks each presented by Henry Kapteyn and Margaret Murnane)
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McElvain Lecture, University of Wisconsin Chemistry Department, February 2012.
Seminar, Margaret Murnane et al, “Attosecond Light and Science - Bright High Harmonic X-Rays at 8Å,” The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA May 2011. Presented by Margaret Murnane.

Number of Presentations: 14.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received		Paper
10/05/2011	4.00	Agnieszka Jaro?-Becker, Craig W. Hogle, Vandana Sharma, Xibin Zhou, Andreas Becker, Henry C. Kapteyn, Margaret M. Murnane, Wen Li. Visualizing Electron Rearrangement in Space and Time during the Transition from a Molecule to Atoms, International Conferece on Ultrafast Phenomena 17. 2011/07/22 02:00:00, . : ,
10/05/2011	5.00	P Ranitovic, X.M. Tong, C W Hogle, X Zhou, M M Murnane, H C Kapteyn. Control of Absorption Cross-Section in He – Towards the XUV/IR-Induced Transparency by the Interference of Electronic Wave Packets, International Conference on Ultrafast Phenomena 17. 2010/07/22 02:00:00, . : ,
TOTAL:	**2**	

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

2012 Appointed Chair of the President's Committee for the National Medal of Science (Murnane)
2012 Outstanding Physics Department Graduate, Leigh Martin (undergraduate student)

2012 Willis Lamb Award for Laser Science and Quantum Optics (Kapteyn and Murnane)
2010 NSF Graduate Fellowship (Daniel Hickstein - student) ?
2010 Appointed to the President's Committee for the National Medal of Science (Murnane)
2010 R.W. Wood Prize of the Optical Society of America (Murnane and Kapteyn)
2010 Schawlow Prize in Laser Science of the American Physical Society (Kapteyn and Murnane)
2009 Ahmed Zewail Award of the American Chemical Society (Kapteyn and Murnane)
2009 NSSEFF DOD Faculty Fellowship for X-ray Generation (Murnane)

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Trevor Courtney	1.00	
Craig Hogle	1.00	
William Peters	1.00	
Daniel Hickstein	0.20	
Chengyuan Ding	0.50	
FTE Equivalent:	**3.70**	
Total Number:	**5**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Henry Kapteyn	0.00	Yes
Margaret Murnane	0.00	Yes
David Jonas	0.00	Yes
FTE Equivalent:	**0.00**	
Total Number:	**3**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED	Discipline
Leigh Martin	0.00	Physics
FTE Equivalent:	**0.00**	
Total Number:	**1**	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period:	1.00
The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:	1.00
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:	1.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):	1.00
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:	0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense	0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:	0.00

Names of Personnel receiving masters degrees

NAME
Total Number:

Names of personnel receiving PHDs

NAME
Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Technology Transfer

FINAL PROGRESS REPORT TEXT

Period covered by report: August 1, 2010 to 8/18/2012

Proposal Title: Bright Coherent Optical Waveforms from the Infrared to the Vacuum Ultraviolet for Manipulation and Detection of Molecules

Contract/Grant number: W911NF-08-1-0391

ARO Program Manager: DOD Army ARO, Richard.Hammond@us.army.mil

Author(s) of report: Margaret Murnane, David Jonas and Henry Kapteyn

Performing Organization Name(s) and Address(es): JILA, Department of Physics, Department of Chemistry, University of Colorado at Boulder, Boulder, CO 80309

ARO proposal number: 54364-PH

People supported:

(2) Student/Supported Personnel Metrics **for this Reporting Period** (name, % supported, %Full Time Equivalent (FTE) support provided by this agreement, and total for each category):

(a) Graduate Students:
Courtney Trevor (100% August 2010), Craig Hogle (100% August 2010-August 2012), Dan Hickstein (working on project from start, supported by NSF Fellowship), William Peters (100% May 2011-August 2012), Chengyuan Ding (50% Jan 2010-May 2011)

(b) Post Doctorates: none

(c) Faculty: Margaret Murnane, David Jonas and Henry Kapteyn, no support

(d) Undergraduate Students: Leigh Martin did his undergraduate honors thesis on this project (unpaid).

(e) Graduating Undergraduate Metrics (funded by this agreement and graduating during this reporting period):
i. Number who graduated during this period : 1
ii. Number who graduated during this period with a degree in science, mathematics, engineering, or technology fields : 1
iii. Number who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields: 1
iv. Number who achieved a 3.5 GPA to 4.0 (4.0 max scale); 1
v. Number funded by a DoD funded Center of Excellence grant for Education, Research and Engineering: 0
vi. Number who intend to work for the Department of Defense:
vii. Number who will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields

(f) Masters Degrees Awarded (Name of each, Total #): None

(g) Ph.D.s Awarded (Name of each, Total #): None

(h) Other Research staff (Name of each, FTE % Supported for each, Total % Supported):

None

Scientific Progress and Accomplishments (description should include significant theoretical or experimental advances)

Four exciting advances resulted from this award –

1. Higher energy ultrafast pulses in the vacuum to extreme ultraviolet (5 – 60 eV)

We recently demonstrated experimentally and theoretically how to substantially increase the conversion efficiency of high harmonic generation (HHG) in the vacuum and extreme ultraviolet (VUV and EUV) regions of the spectrum. Prior to our work, scientists had assumed that to generate the brightest harmonics in the VUV region of the spectrum, a two-color driving laser field was required. We showed, however, that the two most important factors for achieving the highest HHG conversion efficiency in a traditional high harmonic generation experiment are:
1) The driving laser wavelength should be as short as possible, while still being able to generate the desired harmonics. The shorter wavelength also corresponds to a shorter laser period, which means that the electron spends less time away from the ion. This reduces quantum diffusion, and therefore enhances HHG emission from each individual atom.
2) The HHG frequency upconversion process must be phase matched, to ensure that the emission from many atoms in the medium adds together coherently.
This new understanding allowed us to achieve record conversion efficiencies for HHG ($>10^{-3}$) in the VUV/EUV regions around 20 – 60 eV, when driven by 400 nm and 267 nm femtosecond pulses from an optical parametric amplifier (OPA). Through these experiments, we were able to validate a complete theoretical understanding of HHG in the VUV region of the spectrum for the first time. However, these initial experiments used a 10 Hz repetition rate Ti:sapphire laser with poor beam quality to pump the OPA. Prior to publication, we want to repeat these experiments using a new 20mJ kHz laser system that has excellent beam quality. This will allow us to further increase the energy per harmonic that we can obtain (to > 1 μJ).

At longer wavelengths in the UV region, we used four-wave mixing in a waveguide to obtain high energy (> 35 μJ) pulses at wavelengths around 267 nm, which compressed to a 12 fs pulse duration. This represents a significant x4 increase in energy compared with what has been demonstrated to date. Although shorter pulses with durations around ≈3-5 fs had been generated by others, their low pulse energy of ≈ 1 μJ limits their use to molecules with very large absorption cross-sections

2. Electromagnetically Induced Transparency in the VUV

In exciting recent work published in Physical Review Letters (and highlighted as an Editor's Choice), we showed for the first time that it is possible to induce electromagnetic transparency in an atom (He) subject to intense vacuum VUV radiation with photon energy hν~17 - 20 eV. Normally, when atoms are irradiated with intense femtosecond laser and VUV fields, they will ionize through multiphoton processes - even if the energy of the VUV photon is below the ionization potential of the atom (the ionization potential of He is 24.6 eV). However, in the presence of two VUV photons of different energies and an intense infrared laser field, the laser field can modify the electronic structure of the atom, while the presence of two different VUV photons can lead to two distinct ionization pathways that can interfere destructively (see Fig. 1). As a result, *multiphoton ionization of an atom can be turned on or off by manipulating these two interfering pathways.*

This work demonstrates a new approach for coherent control in a regime of highly excited states and strong optical fields, extending ideas from the field of quantum control to a new VUV region of the electromagnetic spectrum using novel control schemes. In the two-photon absorption schemes widely used for coherent control using visible laser fields, the phase of a weak, non-

perturbative, IR pulse can control absorption through interfering two-photon transitions. Our multicolor, multiphoton VUV+IR multiphoton ionization scheme is similar in some respects, except that in our case, the final state is in the continuum i.e. the final state is an ion and free electron.

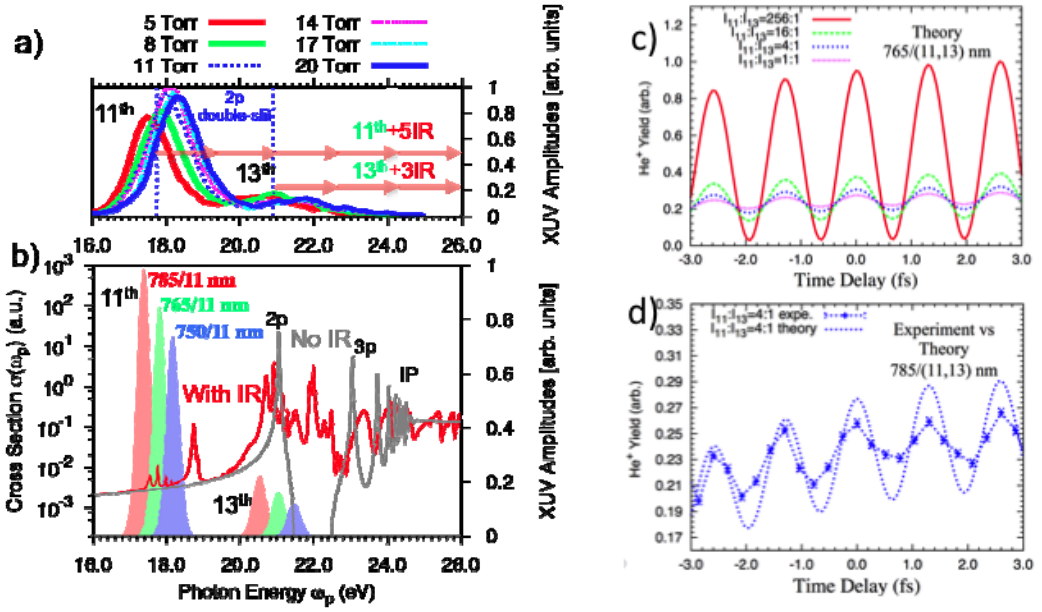

Figure 1: Multiphoton ionization of He in the presence of two interfering quantum pathways: 11ω+5ω and 13ω+3ω. (a) The 11th and 13th harmonics are tuned to the laser-dressed 2p double-slit state by varying the phase matching conditions in a Xe-filled waveguide. The dashed vertical lines show the calculated sidebands of the dressed 2p state of He. (b) Calculated He excitation cross section, with and without the presence of an IR field of intensity 4×10^{12} W/cm^2. By manipulating the absorption cross section of He using the IR laser field, the amplitudes of the two interfering quantum pathways are controlled. (c) Calculated modulation of the He ionization probability for different ratios of the 11th:13th harmonic fields. The red curve demonstrates that for an optimized ratio of 256:1 for the 11th vs. the 13th harmonic amplitudes, nearly complete cancellation of VUV multiphoton ionization can occur. (d) Experimental and theoretical modulation of the He ionization yield for non-optimum intensity ratio of 4:1.

This result was unanticipated because previous work had analyzed multicolor, multiphoton, ionization in combined VUV and IR fields purely in the time domain - not in the frequency domain, where the presence of two interfering channels can be more easily understood. Therefore, past work missed the possibility of manipulating the electronic states in an atom using light in order to control the amplitude and phase of two multiphoton ionization channels.

This approach opens up new possibilities for coherent control of highly excited states, and emphasizes the important and complex role the IR laser field plays in strong field ionization. We believe that this concept can be applied to induce and control the outcome of chemical reactions, as well as in condensed matter physics where the concept of the resonance dressing, shifting and broadening can be applied to the electronic band structure.

3.Capturing multi-electron density in a molecule as a bond breaks

In exciting work published in *PNAS* in late 2010, we used a COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) reaction microscope to visualize both in space and time the dynamic evolution of the entire valence shell electron density as a molecular bond ruptures – a feat that was never before achieved. In this experiment, an ultrashort UV laser pulse was used to initiate dissociation of a Bromine molecule by promoting it into a neutral dissociating state. *The electron density of all 10 bonding electrons was then visualized using an intense IR laser field to ionize the molecule as it fell apart, allowing us to probe electron rearrangement in the entire multi-electron valence shells for the first time.*

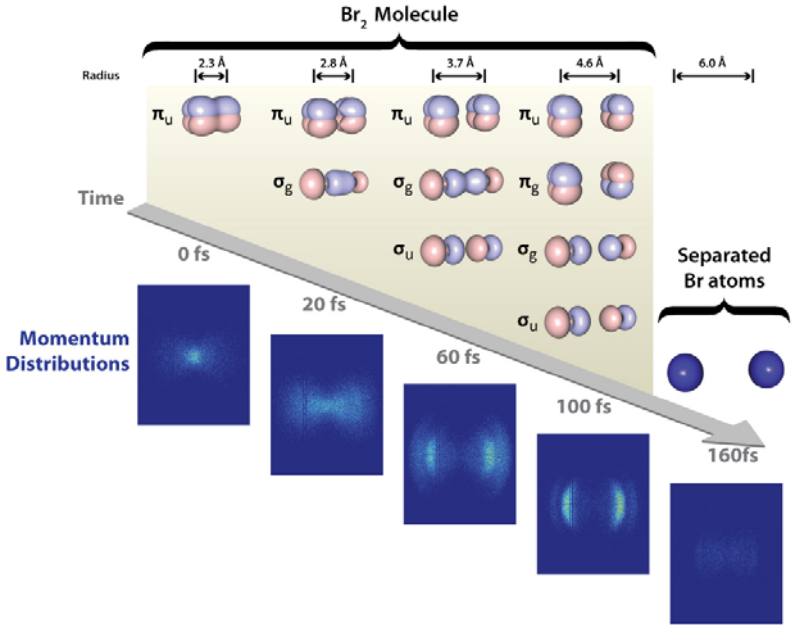

Figure 2: (Top) Calculated molecular orbitals as a function of Br_2 internuclear separation and time after a 400 nm pulse causes the bond to rupture through a dissociating neutral state. All 10 electrons in the valence shell of the molecule are theoretically modeled. (Bottom) Experimentally measured angular-dependence of the Br^+ ion yield from strong field ionization of the Br_2 molecule, as a function of time after the dissociating pulse. Both experiment and theory show that the electron density remains molecular-like for long times, 140fs after the dissociating pulse.

We made a surprising finding illustrated in Fig. 2 – that the system remains molecular-like for a much longer time than had previously been realized, for up to 140 fs after the molecule started to break apart into its components atoms. This means that the electrons do not localize onto the individual atoms until the fragments are quite far apart (> 5 Å), in a region where the potential energy curves for the dissociation are essentially flat, and where there is negligible electron wave function overlap for the two atoms.

In contrast, other approaches that use either photoelectron spectroscopy or high harmonic generation as probes can only capture the dynamics of a single electron as the molecule falls apart into its component atoms. Moreover, these probes appear to be not as sensitive to the electron dynamics in the dissociating molecular system, at least in the experiments performed to date. This situation led to an incorrect estimate of when the electronic density in a molecule becomes atomic-like, that was too short by approximately a factor of two (or ≈ 85 fs). This work highlights the central role of electronic dynamics in a reaction, independent of the nuclear potential energy curves. This work resulted from a collaboration with theorists Agnieszka Jaroń-Becker and Andreas Becker.

4. Spatiotemporal quasi-phase matching

Since its inception by Bloembergen in the early 1960's, quasi phase matching (QPM) was always regarded as a *spatial* method to overcome momentum imbalance between photons participating in a nonlinear optical conversion process. In this work, we theoretically developed a new and generalized realization of quasi phase matching - called spatiotemporal quasi-phase matching - that generalizes traditional *spatial* QPM using layered media to *space-time* QPM.[7] In this theoretical work, illustrated schematically in Fig. 3, we showed that instead of simply being

described by a momentum mismatch, in general a nonlinear optical process may possess *an energy mismatch, a momentum mismatch, or both an energy and momentum mismatch*. In the latter case, any QPM scheme must involve a temporal component.

This new concept of spatio-temporal QPM is quite general and touches on fundamental aspects of all nonlinear frequency conversion processes. Spatio-temporal QPM can be applied to many low- and high-order nonlinear optical processes. In low-order nonlinear optics, spatio-temporal QPM could be used to tune the position of VUV harmonics so that particular molecular spectral features can be accessed. In high order harmonic generation, an accelerating modulation can lead to exquisite control of the high harmonic generation process – leading to the enhancement of a super-wide continuum of harmonics at a predetermined sub-optical cycle of the driving field. This work was published in *Nature Photonics* in 2010.

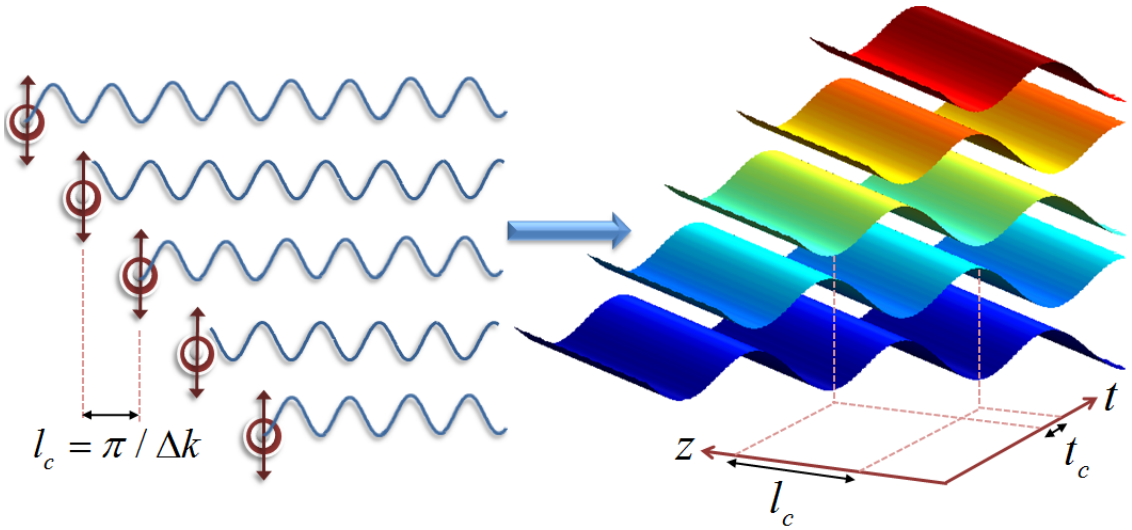

Figure 3. Extending the concept of spatial mismatch to spatio-temporal mismatch. On the left is the traditional graphics used to illustrate the concept of spatial phase mismatch. There, dipole moment radiation of the generated harmonic would be completely out of phase compared to the same radiation emitted a coherence length l_c away. The extension to spatio-temporal mismatch on the right is natural considering that dipole moments radiate in space time, which leads to the possibility that the radiation would be completely out of phase after some distance (the coherence length l_c) and some temporal delay (the coherence time t_c).

Publications from Prior Support

1. A. Bahabad, M.M. Murnane, H.C. Kapteyn, "Quasi Phase Matching of Momentum and Energy in Nonlinear Optical Processes", Nature Photonics **4**, 570 (2010).
2. W. Li, Agnieszka A. Jaron-Becker, C.W. Hogle, V. Sharma, X. Zhou, A. Becker, H.C. Kapteyn, M.M. Murnane, "Visualizing electron rearrangement in space and time during the transition from a molecule to atoms", PNAS **107**, 20219 (2010).
3. P. Ranitovic, X.M. Tong, C. Hogle, X. Zhou, Y. Liu, N. Toshima, M. Murnane, H. Kapteyn, "Controlling the XUV Transparency of He using Two Pathway Quantum Interference," Phys. Rev. Lett. **106**, 193008 (2011) (highlighted as Editor's Choice).
4. W. Li, Agnieszka A. Jaron-Becker, C.W. Hogle, V. Sharma, X. Zhou, A. Becker, H.C. Kapteyn, M.M. Murnane, "Visualizing Electron Rearrangement in Space and Time during the Transition from a Molecule to Atoms," in Ultrafast Phenomena XVII (Oxford University Press, New York, 2011), pp. 83-85.
5. P. Ranitovic, X.M. Tong, C.W Hogle, X. Zhou, M.M Murnane, H.C Kapteyn, "Ultrafast

Modulation of the XUV Absorption Cross-Section of He through Interference," in Ultrafast Phenomena XVII, ed. (Oxford University Press, New York, 2011), pp. 71-73.

Honors during Prior Support

2012 Appointed Chair of the President's Committee for the National Medal of Science (Murnane)

2012 Outstanding Physics Department Graduate, Leigh Martin (undergraduate student)

2012 Willis Lamb Award for Laser Science and Quantum Optics (Kapteyn and Murnane)

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2009 Ahmed Zewail Award of the American Chemical Society (Kapteyn and Murnane)

2009 NSSEFF DOD Faculty Fellowship for X-ray Generation (Murnane)

Invited and Keynote Presentations

Ahmed Zewail Prize talks, Annual Meeting of the American Chemical Society, Salt Lake City, UT March 2009. (Two talks presented by Margaret Murnane and Henry Kapteyn)

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McElvain Lecture, University of Wisconsin Chemistry Department, February 2012.

Seminar, Margaret Murnane et al, "Attosecond Light and Science - Bright High Harmonic X-Rays at 8Å," The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA May 2011. Presented by Margaret Murnane.